

Optimizing the efficiency in high order harmonic generation optimization by two-color fields

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Abstract. We address the problem of the optimization of high-order harmonic generation by two laser fields of frequencies ω_0 and $2\omega_0$ respectively, through a theoretical approach. The parameters that are investigated are the two lasers intensities as well as the relative phase between the two fields. We show that a moderate, but significant increase of efficiency can be achieved in that context and give the domain of optimized parameters. Moreover, we quantitatively investigate the gain and set the limitation of the optimization using such coherent control methods.

PACS. 42.65.Ky Harmonic generation, frequency conversion – 42.79.Nv Optical frequency converters – 32.80.Wr Other multiphoton processes

1 Introduction

For a few years now, high-order harmonic generation (HOHG) has invariably been one of the major topics of super intense laser-atom physics. High order harmonics are generated when a short, intense laser pulse interacts with matter. HOHG has been mainly studied in atomic gases, ions, molecules, atom clusters and solids, and has already been reviewed in several articles [1–4]. Without any doubt, HOHG nowadays stands as one of the most promising methods of producing short-pulse coherent radiations in the XUV range.

In fact, high harmonics have found numerous applications in various areas of physics. More particularly, there have been several applications in atomic physics [5, 6]. Besides, harmonics have already been used for solid state spectroscopy [7], and plasma diagnostics [8]. Applications that are directly related to the coherence properties of harmonics are discussed in reference [4].

As far as the applications of HOHG are concerned, the major problem consists in optimizing various harmonics parameters. Thanks to the improvement of experimental techniques and of theoretical understanding, numerous kinds of optimization of HOHG have been studied. Particularly important are the optimization studies of harmonic efficiency as a function of laser parameters, such as laser polarization, pulse duration, or wavelength [9–12]. Another promising way to improve generation efficiency employs generation by multicolored laser fields, which was

intensively analyzed both theoretically [13–16], and experimentally [13, 17–20].

The evidence of a significant effect of the 2-color mixing on harmonic generation is clear and has been reported in the above cited works. However, nowadays experiments making use of high harmonics (*e.g.* non-linear atomic physics with XUV radiation) require an ever increasing conversion efficiency. Despite the fact that the 2-color non-linear effect might be huge, the question whether such a 2-color technique is able to provide a better conversion efficiency than the classical single laser method has to be discussed and investigated. Thus, the aim of this work is to compare the best harmonic generation rate obtained with a single laser to the best harmonic rate generated with the 2-color scheme, and to conclude whether this technique is worth implementing experimentally.

This quantitative study is carried out within the single active electron approximation by solving the 3D time-dependent Schrödinger equation for an isolated atom in a strong laser field. The interaction with the atom is fully represented without approximations and includes saturation due to ionization. Effects due to the propagation in the medium have been excluded in this initial work. Therefore, the results presented in this paper have to be considered as upper limits since propagation of the harmonics in the gaseous medium will act as a moderator for the optimized conversion efficiency.

The paper is organized as follows. In Section 2, we present our method of solving the time-dependent Schrödinger equation. In Section 3, we discuss how total ionization is affected by a change of the different

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parameters. Sections 4 and 5 contain the main results of our paper, *i.e.* comparative studies of harmonic generation efficiency in single, and two-color laser fields.

2 Numerical simulation

The approach described in this section concerns harmonics generated by an isolated atom subject to the laser pulse. In other words, we limit our study to the Single Atom Response (SAR), and exclude effects due to the propagation of the scattered light in the gaseous medium. Thus our approach involves the solution of the Time-Dependent Schrödinger Equation (TDSE) associated with the atomic system illuminated by a mono- or a bi-chromatic laser pulse. The atomic system we consider is hydrogen and the field is described semi-classically. For the present work, the polarization is set linear. The numerical method we use to solve the TDSE is one of the most precise ones, and has been described elsewhere [21] in the context of the studies of Above-Threshold-Ionization spectra. It is based on the decomposition of the total wave-function onto a basis of spherical harmonics and B-spline radial wave-functions. This technique has also proved to be very well suited for the calculation of HOHG. As in similar approaches [22–24], the information relative to the high order harmonics is directly obtained from the Fourier-transform of the time-dependent electric dipole, or from its acceleration if ionization is important.

Numerical simulation of HOHG is very well documented (for a recent review see [4]). However, several points still need to be clarified, or emphasized in the present context. First of all, the interaction is described in the velocity gauge. This choice is almost a necessity when one uses the wave-function expansion in terms of spherical harmonics, together with intense laser fields [25]. Therefore, the electromagnetic field is defined through the vector potential as:

$$A(t) = \frac{\mathcal{E}_0}{\omega_0} f(t) (\cos(\omega_0 t) + \frac{r}{2} \cos(2\omega_0 t + \phi)), \quad (1)$$

$$E(t) = -\frac{\partial}{\partial t} A(t), \quad (2)$$

where ω_0 is the fundamental laser frequency, \mathcal{E}_0 is the field maximum amplitude with a corresponding intensity $\mathcal{I}_0 = \mathcal{E}_0^2$, r^2 is the intensity ratio between the two field components such that the total intensity is given by $\mathcal{I} = \mathcal{I}_0(1+r^2)$. Finally ϕ is the relative phase. The choice of the pulse shape $f(t)$, which is supposed to mimic the temporal evolution of the laser envelope, is an important issue. A good approximation is obtained with envelope functions of the form: $f(t) = \exp(-(t/\tau)^2/2)$ with $t \in [-\infty, \infty]$ or $f(t) = \cos(\pi t/\tau')$ with $t \in [-\tau'/2, \tau'/2]$. Computing the harmonic spectrum with such pulse shapes where the full width at half maximum does not exceed 100 fs results, however, in an irregular spectrum in which the harmonics located in the plateau region cannot be resolved [26,27]. Note, nevertheless, that such a spectrum clearly exhibits harmonics if propagated through the medium [28]. This

phenomenon is due to the fact that only harmonic frequencies are phase matched in the medium and therefore propagate with a minimum loss. A way to circumvent this problem inherent to the single atom calculations is either to use pulses longer than 100 fs inducing an extra computation costs, or to consider a modified shape such as a “flat pulse” which maintains the field envelop constant over several cycles [29]. We have opted for such pulses which include a cosine squared turn-on and -off. We were able to check that such a “flat pulse” with a FWHM of 50 fs could resolve the plateau harmonics; in contrast, to achieve the same resolution, it was necessary to extend the pure cosine squared pulse up to a FWHM of 200 fs.

Ionization is a relevant physical quantity in the context of HOHG since it acts as an inhibitor: when the atom is totally ionized, the dipole does not radiate anymore. Our numerical approach allows to compute ionization in two different ways which are equivalent in principle, but may be different in practice. The first technique available is to compute, by projection onto the bare atom eigenstates, the populations left in the bound states, as well as in the continuum states at the end of the pulse. Ionization degree is then defined as the complement to the population left in the bound states. The second technique involves an absorbing wall set at the boundaries of the finite box in which the system is described, *i.e.* a sphere in the present case. Every wave-packet released in the continuum by the field, moves away and is absorbed by the wall when it reaches the space boundaries. Since the absorber is modeled by a complex potential, the norm of the total wave-function decreases as the outgoing density probability flux hits the wall. In that case, ionization is defined as the missing part of the norm of the wave-function. The great advantage of the latter method is that relatively small “boxes” are required to represent the interaction, and therefore the computation time remains reasonable. On the other hand, such an approach leads to a computed ionization degree that depends, for a given pulse duration, on the box size. The population that has reached the boundaries during the pulse would never have done so if the box was 10 times bigger (it requires much more time to travel to the far boundaries). However, if one keeps propagating the system after the pulse has gone until all the population in the continuum has flown away (absorbed by the wall) for any box size, then the ionization degree computed in this way is independent of the box size, and equals that computed by the projection technique. The projection method has been implemented in this work.

3 Total ionization

Harmonic generation is a highly non-linear process, and as such is very sensitive to the field intensity. Increasing the field strength has mainly two effects. First, it moves the position of the cut-off towards lower wavelengths, thus extending the plateau. Second, the harmonic strength itself increases. However these processes are subject to saturation when the atom has completely ionized. Above the saturation intensity, any further increase of the intensity

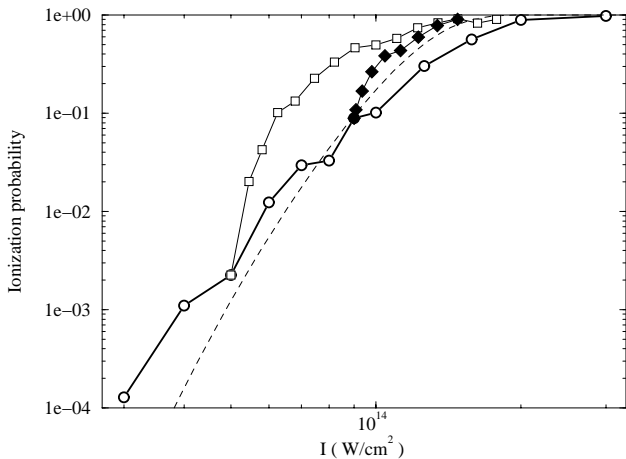


Fig. 1. Ionization of H as a function of the two-color field intensity ($\hbar\omega_0 = 1.33$ eV, FWHM = 50 fs, $\phi = 0$). Open circles: ω_0 alone, open squares: ω_0 and $2\omega_0$ $\mathcal{I}_0 = 5 \times 10^{13}$ W/cm² and r varies from 30 to 160%, filled diamonds: ω_0 and $2\omega_0$ $\mathcal{I}_0 = 9 \times 10^{13}$ W/cm² and r varies from 10 to 80%, dashed line: ADK calculation.

results in a less efficient harmonic generation. Ionization is therefore a key point as a limiting mechanism in HOHG. Moreover, the highest conversion efficiency is observed close to the saturation intensity. It is then crucial to study how the combination of the two fields acts on total ionization. We present in this section the variations of the ionization probability with respect to several parameters of the fields (\mathcal{I}_0 , r).

A complete study would consist in computing harmonic generation due to the bichromatic field for all possible values of the intensity of each frequency, that is scanning over the two parameters \mathcal{I}_0 and r . Such a task may result in a quite heavy computation. We have therefore restricted ourselves to considering only two cases *i.e.* a study as a function of the relative intensity r^2 for two representative intensities of the fundamental frequency ($\hbar\omega_0 = 1.33$ eV), namely $\mathcal{I}_0 = 5 \times 10^{13}$ W/cm² (low ionization when $r = 0$), and $\mathcal{I}_0 = 9 \times 10^{13}$ W/cm² (close to saturation already when $r = 0$). Figure 1 shows the total ionization probability due to the fundamental frequency alone, and to the mixed two color field. One immediately sees that the presence of the $2\omega_0$ field greatly enhances ionization, and that saturation is reached with much lower intensities. The explanation is straightforward. It takes 11 photons to ionize the atom with the single fundamental field. Therefore, within the lowest order perturbation theory, the probability varies like \mathcal{I}_0^{11} . On the other hand it takes only 6 photons to ionize with the $2\omega_0$ field. The order of the process is thus almost twice as less, and therefore more efficient at comparable intensity. For comparison, we also show in Figure 1 the ionization degree due to the fundamental field calculated using the ADK model [30]. As expected, the agreement between exact numerical and ADK results becomes worse as the intensity decreases entering the multiphotonic regime (the Keldish parameter being $\gamma = 1.45$ at $\mathcal{I}_0 = 4 \times 10^{13}$ W/cm²).

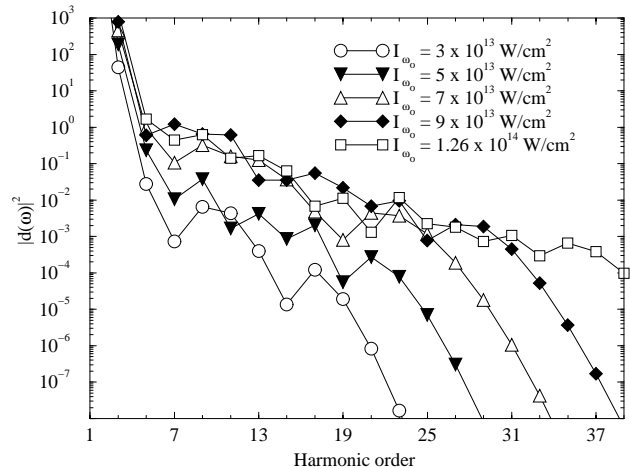


Fig. 2. Harmonic spectra of H as a function of the single field intensity ($\hbar\omega_0 = 1.33$ eV, FWHM = 50 fs).

The addition of the $2\omega_0$ component might help improving HOHG, but certainly enhances ionization lowering the saturation intensity.

4 Harmonic generation due to the single frequency field

Before going any further in investigating the response of the atom to the superposition of the two components ω_0 and $2\omega_0$ of the bichromatic field, we present typical harmonic spectra due to either the ω_0 or $2\omega_0$ component alone. We will also determine the optimum parameters producing the maximum number of harmonic photons.

First of all, let us recall the general behavior of the harmonic spectrum as the intensity is varied. One easily sees in Figure 2 that increasing the intensity results in a better harmonic generation efficiency, regardless of the order. However, it should be noticed that above a certain intensity, this efficiency decreases due to the depletion of the atom (see Sect. 3). To summarize, the harmonic photon number varies as follows: it increases very rapidly until it reaches the plateau region, then changes levels off with wide oscillations over a certain intensity range, and plummets when the saturation intensity is exceeded. This behavior is illustrated in our case for three selected harmonics, namely H11, H21 and H33, in Figure 3. The optimal regime is determined by selecting the very intensity for which a given harmonic shows the maximum strength.

This behavior is obviously similar to the case where the fundamental field has the $2\omega_0$ frequency. The only difference being that an extended plateau does not have a chance to build up before the atom has ionized, the ionization process is much more efficient in that case. However, if this frequency does not allow for high order harmonics, the strengths of the low order harmonics produced (from H3 to H11 of the $2\omega_0$ field) are much larger than those obtained with the ω_0 field [18], as it can be seen in Figure 4. Harmonics of the $2\omega_0$ field are displayed in Figure 4 as a function of the fundamental field harmonic

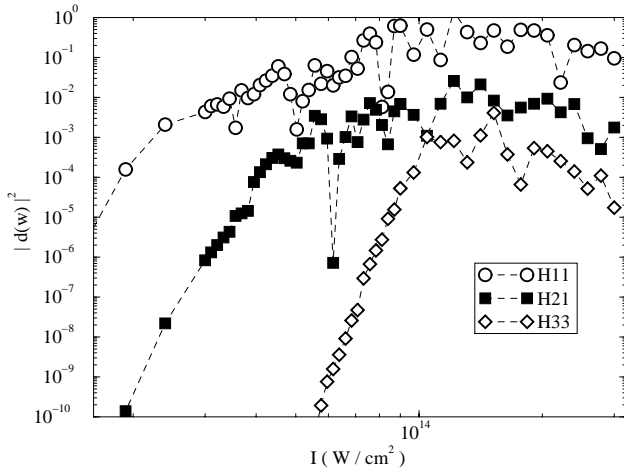


Fig. 3. Variation of the polarizability of H11, H21, H33 as a function of the laser intensity ($\hbar\omega_0 = 1.33$ eV, FWHM = 50 fs).

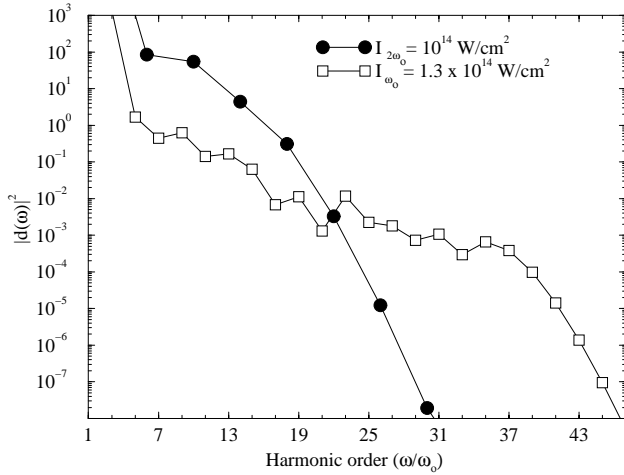


Fig. 4. Optimal spectra obtained with ω_0 alone and $2\omega_0$ alone ($\hbar\omega_0 = 1.33$ eV, FWHM = 50 fs).

order, and therefore appear at even values of the abscissa. This higher conversion efficiency is explained in terms of wave-packet diffusion [31]. HOHG is based on the recollision and recombination of an oscillating freed electron with its parent ion. In the presence of the $2\omega_0$ field, the time a driven continuum wave-packet needs to come back to the nucleus is half as long as that in the presence of the ω_0 field. Therefore, the spreading of the wave-packet is less important, and results in a larger recombination cross-section.

Figure 4 shows the best achievable spectra (in terms of the photon number) due to either of the two fields. The message to be read from this graph is the following: as long as one is interested in photons with energy less than $21\omega_0$ it is preferable to generate the radiation from the $2\omega_0$ field alone for a better efficiency. Note however that the number of harmonic frequencies available is much less in that case due to the energy spacing of $4\omega_0$ between consecutive harmonics. On the other hand, larger photon energy is only available *via* the interaction with the ω_0 field alone. An attempt to enhance HOHG by mixing the

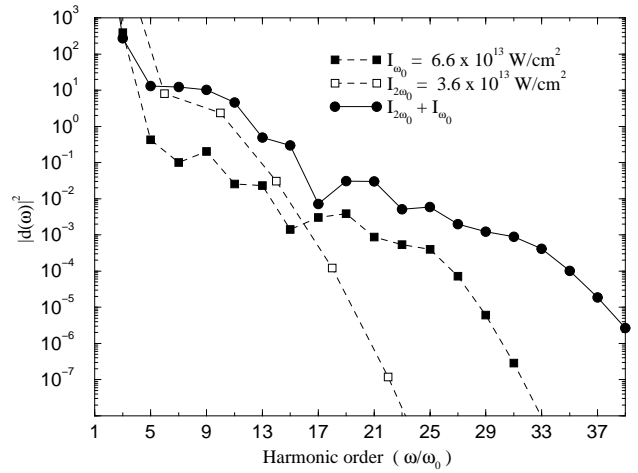


Fig. 5. Comparison of the spectrum of odd harmonics produced by the bichromatic field and those obtained using each field component individually ($\hbar\omega_0 = 1.33$ eV, FWHM = 50 fs, $\phi = 0$).

two frequencies has to provide spectra that are better (*i.e.* have higher efficiency) than the maximum of the joined spectra of Figure 4.

5 Harmonic generation in the bichromatic field

The aim of this section is first to show that the effect of the mixing on harmonic enhancement can be very large but not necessarily large enough to provide significant gain in the harmonic production compared to what can be obtained using a monochromatic field.

As it has been already reported in both experimental as well as theoretical works [17–19,32], the addition to the fundamental field of only few percents of its second, or third harmonic may greatly enhance harmonic generation up to a factor of 10 in the most favorable cases. We also have observed such an effect as it is illustrated in Figure 5, where we compare the spectrum obtained with the ω_0 field alone and the one given by the bichromatic field where the intensity of the $2\omega_0$ component is 50% of the fundamental intensity. The plateau due to the bichromatic field clearly lies between 1 and 2 orders of magnitude above the plateau due to ω_0 component alone. Moreover, the plateau has been extended and the position of the cut-off is now located around H31 (instead of H25 in the single component case). This latter point can be explained by reconsidering the classical rescattering picture leading to the so-called cut-off law [33,34], in the case of the bichromatic field. The kinetic energy at the time, when the electron backscatters with its parent ion, may be larger than the conventional $3.17U_p$ depending on the relative phase between the two field components [35]. This supports the conclusions of a recent analogous work [36] performed in 1D that shows that there is no analog of a simple cutoff law for bichromatic fields.

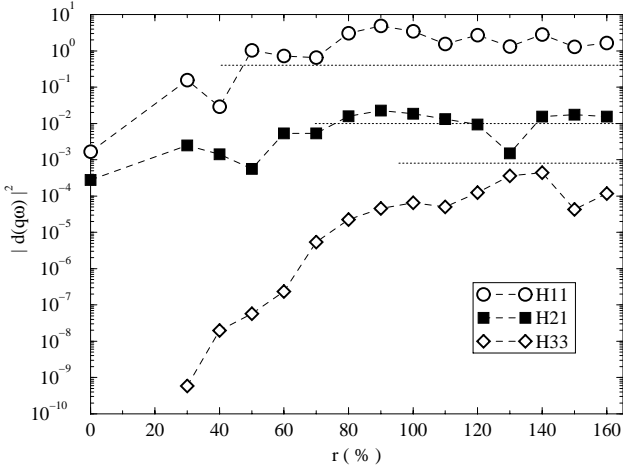


Fig. 6. Polarizability of odd harmonics H11, H21 and H33 for various values of the intensity ratio r . ω_0 -field intensity is 5×10^{13} W/cm². Horizontal dashed lines refer to the averaged maximal corresponding polarizabilities obtained using the ω_0 field alone ($\hbar\omega_0 = 1.33$ eV, FWHM = 50 fs, $\phi = 0$).

Although the effect of the mixing of the two fields (ω_0 and $2\omega_0$) on conversion efficiency is clear and can be large depending on the intensities considered, its existence does not necessarily imply that, given an atomic system, a larger number of harmonic photons can be collected using a mixed field compared to what is commonly achieved with a single monochromatic field. To clarify this point we have performed a quantitative and systematic study of the harmonic production efficiency due to the mixed field for a large set of parameters (intensities of both field components). We then have compared the best results (in terms of photon number) to the harmonic production achievable with the monochromatic field.

For simplicity's sake, we have chosen to illustrate the investigation on a selected set of harmonics, namely H11 (beginning of the plateau), H21 (middle part of the plateau) and H33 (close to the cut-off). We have computed the single atom response at those frequencies as a function of the intensity of each of the two field components. The polarizabilities are plotted in Figure 6 (low fundamental field intensity), Figure 7 (moderate intensity) and Figure 8 (close to the saturation intensity). We have extracted from the curves plotted in Figure 3 the maximum polarizability for each of the 3 harmonics (all the curves have first been smoothed with respect to the wide fluctuations, which in a sense mimics the macroscopic propagation effects [4]). The values thus obtained (0.4 for H11, 0.01 for H21 and 8×10^{-4} for H33) are reported as horizontal dashed lines in Figures 6 to 8. A gain in a given harmonic production due to the mixing of the 2 frequencies is observed as soon as the curve lies above the corresponding threshold (dashed line). A rapid overview of the 3 graphs shows some gain, at least for harmonics H11 and H21, over a significant range of the intensity ratio. This gain can be as big as an order of magnitude (for H11 for example). The behavior of the curves, however, is different for the three intensities considered. As one can see in Figure 8 (high ω_0 -field inten-

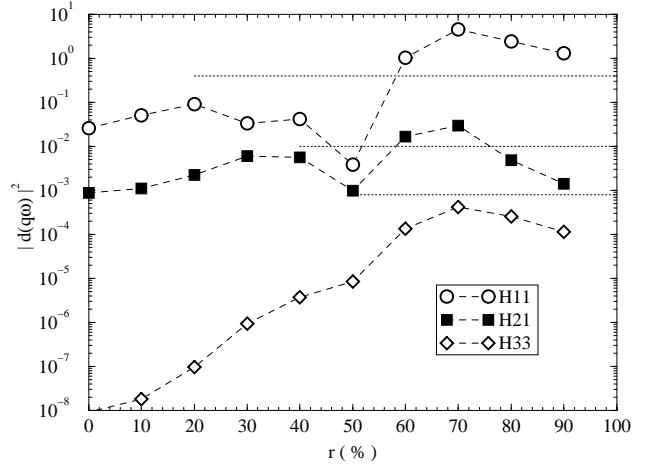


Fig. 7. Same as Figure 6. ω_0 -field intensity is 6.6×10^{13} W/cm².

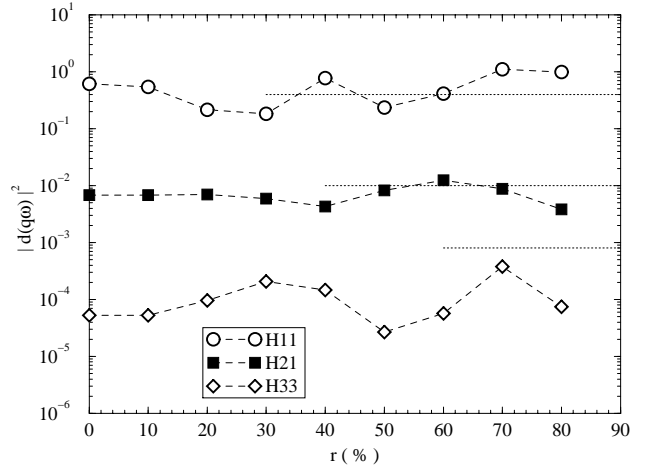


Fig. 8. Same as Figure 6. ω_0 -field intensity is 9×10^{13} W/cm².

sity), the addition of the $2\omega_0$ -field does not enhance significantly the harmonic generation. The curves related to H11 and H21 barely fluctuate around the value obtained in the absence of the $2\omega_0$ field (namely at $r = 0$ in the Figure). Only H33 exhibits an increase around $r = 70\%$ but does not reach the maximum strength observed in the monochromatic case. The situation is, however, different in Figure 7 (moderate ω_0 field intensity) where the curves clearly increase as r increases. Starting around $r = 60\%$, the curves lie above their corresponding threshold (except H33). In fact, although not displayed in the figure, H5 up to H23 are significantly enhanced with respect to the best case obtained with the monochromatic field. What should be remembered here is that the mixing of ω_0 and $2\omega_0$ ($I_{\omega_0} = 6.6 \times 10^{13}$ W/cm², $r = 70\%$) results in the production of a larger number of photons in the harmonic modes H5 up to H23 than the number that can be achieved in the most favorable case ($I_{\omega_0} = 10^{14}$ W/cm²) using the monochromatic field alone. Finally, Figure 6 (low fundamental field intensity) also shows a gain for harmonics lower than H23. The gain looks less spectacular than in the previous case, but has the advantage of being quasi constant (small fluctuations) over a large set of the $2\omega_0$

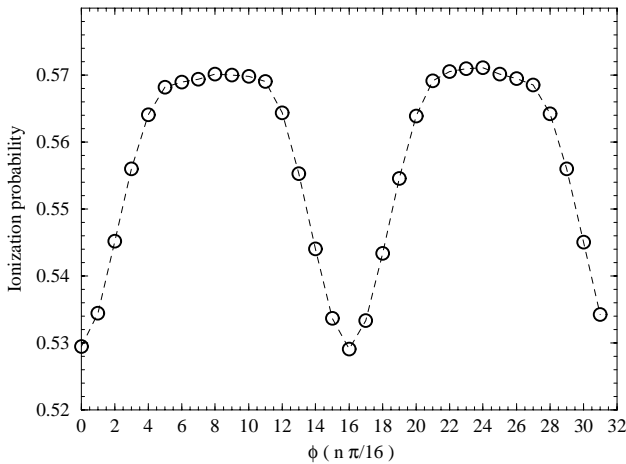


Fig. 9. Ionization as a function of the relative phase. $\hbar\omega_0 = 1.33$ eV, FWHM = 50 fs, $I_0 = 6.6 \times 10^{13}$ W/cm² and $r = 70\%$.

field intensity. In this case, the optimal intensity ratio is somewhere around $r = 100\%$.

So far, we have only considered the two field component intensities as tunable parameters. However, the relative phase between the two fields affects significantly the ionization degree [19,37], and the harmonic strength [15,17,32]. We now propose to determine up to what degree the relative phase affects HOHG in the case where $I_{\omega_0} = 6.6 \times 10^{13}$ W/cm² and $r = 70\%$ since it corresponds to a quite favorable set of parameters as we have shown above. Figure 9 shows the total ionization as a function of the fields phase difference ϕ . As can be shown, ionization is periodic with a periodicity of $2\pi/n$ where n is the ratio of the frequency of the two fields ($n = 2$ in our case). Changing ϕ alters the maximum amplitude of the electric field, although the averaged intensity remains the same (see, *e.g.* Fig. 1 of [37]). Because ionization (multiphoton ionization in our case) is a non-linear process, the phase configuration for which the field is the less symmetric with highest maximum amplitude ($\phi = \pi/2$) leads to a more efficient ionization mechanism as can be seen in Figure 9. Note that in the case $r = 70\%$, the contrast is 0.036, while it is 0.34 for $r = 30\%$. This latter point is in agreement with the general feature according to which the effect of the second harmonic field is less and less significant as the system reaches the saturation regime.

We also have looked at the modulation of the harmonic strength as a function of the phase difference. The results are presented in Figure 10 for a selected set of harmonics. The modulation can be as big as 3 orders of magnitude for a cut-off harmonic, that is, H33 can completely disappear if the phase difference is chosen to be $\pi/2$ for instance. Low order harmonics (up to H5) as well as cut-off harmonics have a similar and well defined behavior, namely they are maximum for $\phi = 0 \pmod{\pi}$ and minimum for $\phi = \pi/2 \pmod{\pi}$. Note that the generation of these harmonics is maximal when ionization is minimal, and *vice versa*. However, the plateau harmonics have a more complex phase dependence. It may happen that a particular harmonic has a maximum strength at non-

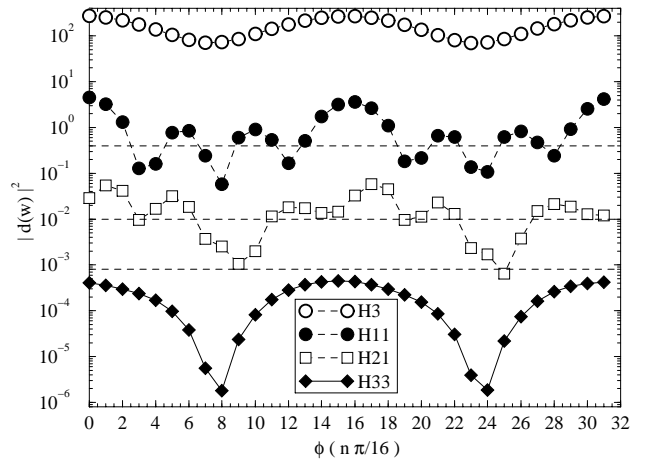


Fig. 10. Harmonic strength as a function of the relative phase. $\hbar\omega_0 = 1.33$ eV, FWHM = 50 fs, $I_0 = 6.6 \times 10^{13}$ W/cm² and $r = 70\%$.

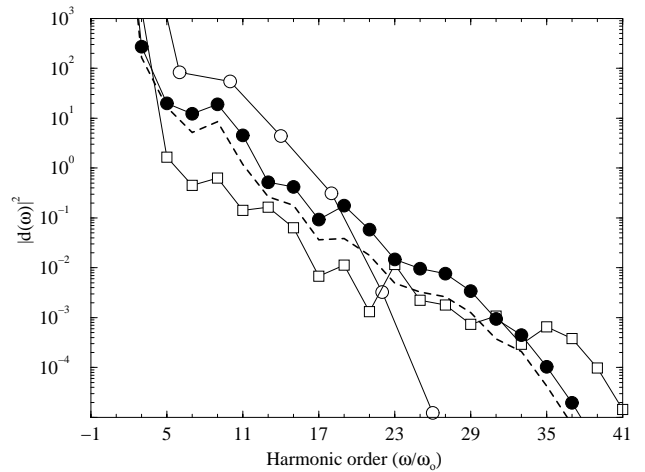


Fig. 11. Open circles and squares: same as Figure 4, filled circles: phase optimized spectrum for $I_0 = 6.6 \times 10^{13}$ W/cm² and $r = 70\%$ ($\hbar\omega_0 = 1.33$ eV, FWHM = 50 fs), dashed line: phase averaged spectrum.

singular values of the phase as it is the case for H21 for example. This is the reason why one should also tune the phase difference in order to optimize the number of photons in each harmonic mode. Note, however, that each mode may have an optimum parameter set (\mathcal{I}_0, r, ϕ), not necessarily identical for all modes. This latter point has been recently studied in another work [20].

Finally, Figure 11 shows the best harmonic spectrum obtained so far, that is, optimized over the field intensities but also over the relative phase.

One can see that the optimized curve (filled circles) lies above the optimal single color spectrum up to harmonic 23 by about one order of magnitude. After that limit, more or less located in the middle of the plateau, the enhancement is not as spectacular but still significant. The signal dies out definitively around H35. Note however, that up to H17 the optimized spectrum lies below the spectrum obtained using the $2\omega_0$ -field alone. The phase-optimized

spectrum has been constructed by collecting the polarizabilities corresponding to the best phase configuration for each individual harmonic. As mentioned earlier, the optimal phase tuning is typically different for different harmonic orders. Also shown, is the spectrum obtained by averaging over the relative phase to evaluate the effect of a non-uniform relative phase distribution over the interaction volume. This case thus sets a lower limit for the investigated phenomena. A reasonable prediction should lie in between these two curves (dashed and filled circles of Fig. 11).

6 Conclusion

We have theoretically investigated harmonic generation using two-color pulsed fields. We have shown that an enhancement of harmonic efficiency can be achieved using frequency mixing, compared to what is obtained using a single field. Enhancement requires, however, that all of the parameters be optimized for a maximal harmonic strength production. This study gives ranges of parameters, for which frequency mixing can be useful (at least for harmonics expanding up to the middle of the corresponding optimized single field harmonic spectrum), and points out that the relative phase between two color field needs also to be optimized. The maximum gain so far achieved sets an upper limit to the maximum enhancement observable in the single active electron approximation. This limit will obviously be lowered by the phase-mismatch conditions appearing during the propagation in the medium. The quantitative evaluation of the effect of absorption and phase-matching on conversion efficiency in such a scheme is numerically involved although within reach on nowadays' super computers and will be considered in view of the preliminary results of a 2-color experiment that will be held in CELIA (Bordeaux, France).

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